Measurement of the Pressure Derivatives of Elastic Constants Using Resonance Ultrasound Spectroscopy

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Progress Report for March 1, 1994–February 28, 1995

ONR Grant N00014–93–0544

In our last year's proposal to the Physical Acoustics Section of the Physics Division of ONR, we outlined an overall goal for three years and a goal for the first year's work. The overall goal was to measure the third order elastic constants of non-isotropic solids. The first year's goal was to "prove out the measuring system and complete the measurements of third order constants on one solid, which will probably be KCl." We also promised to design the equipment to measure the remaining third order constants, not found from the hydrostatic pressure experiment.

The measurement of resonant frequencies under hydrostatic pressure imposes some severe restraints on the measuring apparatus. Restraints arise because pressure will tend to damp the resonant modes, and if sufficient pressure exists, the observed modes will disappear in the noise of the system. We are obliged, therefore, to do the measurements at modest pressure (100–200 bars), which requires that the pressure measuring apparatus be very accurate. We chose the ultimate in accuracy for pressure-generating equipment, the dead weight loader. As a consequence, we had to design and order some specialized equipment. The equipment was purchased early, but when it first arrived it had to be returned to the manufacturer for modification. Our final system arrived considerably behind schedule, which frustrated our attempt to "prove our measuring system" early in the year. We have just completed the first few runs on our system, and have proved out the essential parts of the system. But this delay has prevented us from completing the measurement of third order constants on any solid. We are about to undertake these measurements at the beginning of the second year.

While waiting for the equipment to arrive, we decided to use our time to do some experiments and theory not described in our proposal, but which are essential to the ultimate success of the three year goals. Our experimental sample will be small, so we decided to see whether the elastic moduli, C_{ij}, as determined from the measured resonant frequencies, were independent of the size of the sample. Some experimentalists, when confronted with the observation that the elastic moduli for some materials measured by RUS are higher than those measured by the ultrasonic pulse method, attributed the difference to size effects in the measured results of the RUS technique. We, therefore, cut five parallelepipeds of different sizes from a high quality boule of fused quartz for our measurements. Fused silica was chosen because it is readily available and inhomogeneities across a large specimen are minimal. The specimen sizes varied from 44.5 to 1.5 mm^3. We found no systematic changes in any of the C_{ij} as the size was reduced (Figure 1). We will publish this result soon, with our ONR grant emphasized in the acknowledgements.

We also wished to ascertain whether the C_{ij} measured by RUS were comparable to those measured by other ultrasound techniques; in particular, pulse superposition acoustics and Brillouin scattering. Don Isaak, in combination with Earl Graham at Penn State (pulse

superposition acoustics) and Jay D. Bass at the University of Illinois, Urbana (Brillouin scattering), compared data on Fe_2SiO_4 (fayalite). Two of these investigators (DI and EG) measured the C_{ij} of the same fayalite specimen. The study was completed, and a paper was produced (see references). D. Isaak's participation in these analyses was supported by the ONR grant. This was a landmark paper because reasons for different measured C_{ij} values of fayalite are clarified. Differences in the C_{ij} and $\partial C_{ij}/\partial T$ can be attributed to specimen variations rather than differences produced by the methods of measurement. Of special interest is the observation that ultrasonic pulse-echo and RUS experiments on the same specimen can produce different results in the event of significant specimen inhomogeneities. This result comes about because different parts of the specimen are sampled during these two types of experiments. However, in a homogeneous specimen, we are now assured that a good ultrasonic pulse-echo experiment produces results in good agreement with RUS.

A further set of experiments is to examine the effect on the elastic moduli of change of Fe in the MgO-Fe solid solution. These experiments are being done at several temperatures. The object is to see whether the value of the bulk modulus is sensitive to concentration of the solute. A further objective of these experiments is to investigate the unusual behavior of the C_{44} modulus of FeO at elevated temperature and/or pressure. Earlier pressure experiments show that C_{44} decreases with increased pressure, a very unexpected result, since C_{44} goes up with pressure for MgO. We wish to document the effect of temperature on the C_{44} modulus of FeO (as well as confirm the unusual pressure effect) and assess to what extent this result could have been predicted by application of the equation relating the pressure derivatives of the elastic moduli to the temperature derivatives provided later in this report. A preliminary set of experiments on pure iron has already been completed (see references).

On the theoretical side, we pursued work in establishing a theory for the change of thermal expansion coefficient, α, with T and P. While the change in α will be small in our experiments, it must, nevertheless, be accurately known if we are to establish third order elastic constants and their temperature derivatives. We will have to deal with solids with rather high ambient values of α, and we will need to quantify the change in α due to pressure and temperature, and how this affects the determination of dC_{ij}/dP.

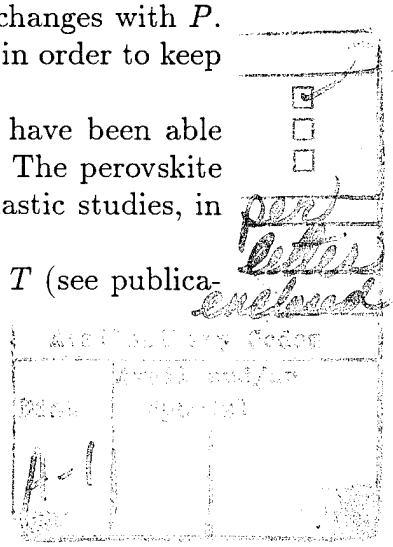
The pressure effect of α is given by the well known thermodynamic identity,

$$B_T^2 \left(\frac{\partial \alpha}{\partial P}\right)_T \equiv \left(\frac{\partial B}{\partial T}\right)_P ,$$

where B_T is the isothermal bulk modulus. Here we see the importance of the temperature experiments in RUS. The measurement of the C_{ij} with T tells us how α changes with P. Further changes of α with P tells us how sensitively T must be controlled in order to keep a constant volume (or find the corrections therefrom).

By developing the necessary equations such as described above, we have been able to determine how α changes with P and T for the perovskite structure. The perovskite structure is important in high T superconductivity studies and in ferroelastic studies, in addition to geophysics studies.

We have published one paper on α of perovskite at high P and high T (see publica-



tions). We intend to use this knowledge in future work in the ONR grant when we measure the third order constants of a perovskite high-T superconductor.

One paper is essentially written, but we are waiting for experimental results to verify certain points before we submit it. It turns out that the variation of any C_{ij} with pressure can be predicted by measuring the variation of that same C_{ij} with temperature: the equation we derived, but which must be verified, is

$$\left(\frac{\partial C_{ij}}{\partial P}\right)_T = -\left(\frac{1}{\alpha B_T}\right)\left(\frac{\partial C_{ij}}{\partial T}\right)_P .$$

We have tested it with data from other laboratories. A better test of accuracy can be made when the pressure experiment is done on precisely the same sample and using the same technique as the temperature experiment. To verify the above expression will be of enormous value to our project because the third order constants we measure can be independently confirmed.

Published Papers Supported by ONR Grant

1. The elastic properties of single-crystal fayalite as determined by dynamical measurement techniques, D.G. Isaak, E.K. Graham, J. Bass, and H. Wang, *Pure Appl. Geophys.*, *141*, 393–414, 1993
2. A thermodynamic method for computing thermal expansivity, α, versus T along isobars for silicate perovskite, O.L. Anderson and K. Masuda, *Phys. Earth Planet. Inter.*, *85*, 227–236, 1994

Papers in Press

3. The thermodynamic route to finding α and other physical properties at high P and high T, O.L. Anderson, K. Masuda, and D.G. Isaak, *Phys. Earth Planet. Inter.*
4. Elastic and viscoelastic properties of α-iron at high temperature, D.G. Isaak and K. Masuda, *J. Geophys. Res.*

Papers Being Written

5. On the calculation of dC_{ij}/dP from measurements of dC_{ij}/dT, O.L. Anderson and D.G. Isaak, intended for *J. Phys. Chem. Solids*
6. Elastic constants of the iron-rich side of the solid solution MgO-FeO, D.G. Isaak and O.L. Anderson, intended for *J. Appl. Phys.*

Appendix

Progress in Setting up the Experimental Apparatus

1. Finished Mounting and Assembling the Dead Weight Tester

We completed the assembly of the pressure system during the past year (Figure 2 a,b). The dead weight tester (DWT) was received from the Harwood Company in January 1994.

Since that time, we have mounted the system in our lab and have assembled it to the extent that we have been able to perform several pressure tests to characterize the relationship between the chamber and jacket pressure (see #2 below). We were unable to couple the system to our pressure vessel (which houses the specimen) because it was only recently, i.e., January 30, 1995, that we were able to obtain the differential pressure cell from the vendor (a 12 month delay in delivery).

2. Completed Testing of High Pressure Vessel/Jacket Pressuring System

A series of tests has been performed to establish the relationships between the DWT chamber pressure, the piston fall time, and the jacket pressure. These relationships are crucial because our experiments require time for temporal thermal gradients to diminish once pressure has been applied. We wish to hold the pressure in the vessel constant for about 30–60 minutes after applying pressure before we obtain resonance data. Constant pressure is maintained if the piston is free to fall in the cylinder. However, if the piston-cylinder clearance is excessive, the piston drop rate will be faster than is required to complete the experiment. The piston-cylinder clearance and, therefore, the piston drop rate, is controlled by applying a jacket pressure to the vessel. We measured the piston drop rate over chamber pressures of 0–120 bar so as to determine the optimal jacket pressure at several values for the chamber pressure. It should be noted that the correct jacket pressure must be determined accurately. The chamber pressure will be erroneously low in the event of too great a jacket pressure being applied, since there will be significant friction between the piston and cylinder.

3. Finished Constructing and Testing Sample Holding Apparatus for Insertion Into the High Pressure Chamber

We obtained a pressure vessel from a vendor in December 1993. During the past year we have completed the construction of an add-on to this vessel on which the specimen is mounted and inserted into the pressure vessel (Figure 2c). We have also testing this mounting/holding device at ambient conditions using a KCl specimen we expect to measure at elevated pressure. An output of the spectrum obtained from this test is seen in Figure 3. At present we are assembling and calibrating the thermocouple pairs that are also mounted to this device.

4. Addition of the Differential Pressure Cell

We were unable to obtain a spectrum at elevated pressure because of the tardiness with which our vendor supplied the differential pressure cell (DPC). The DPC is required to isolate the gaseous fluid in the pressure vessel from the liquid pressure medium (a monoplex and engine oil mixture) in the chamber of the dead weight tester while ensuring that the pressures in the vessel and the chamber of the dead weight tester are equal. We received the DPC in late January, 1995, and are in the process of installing it into our system (Figure 2a,b). The inclusion of the DPC will enable us to begin making spectral measurements at elevated pressure shortly.

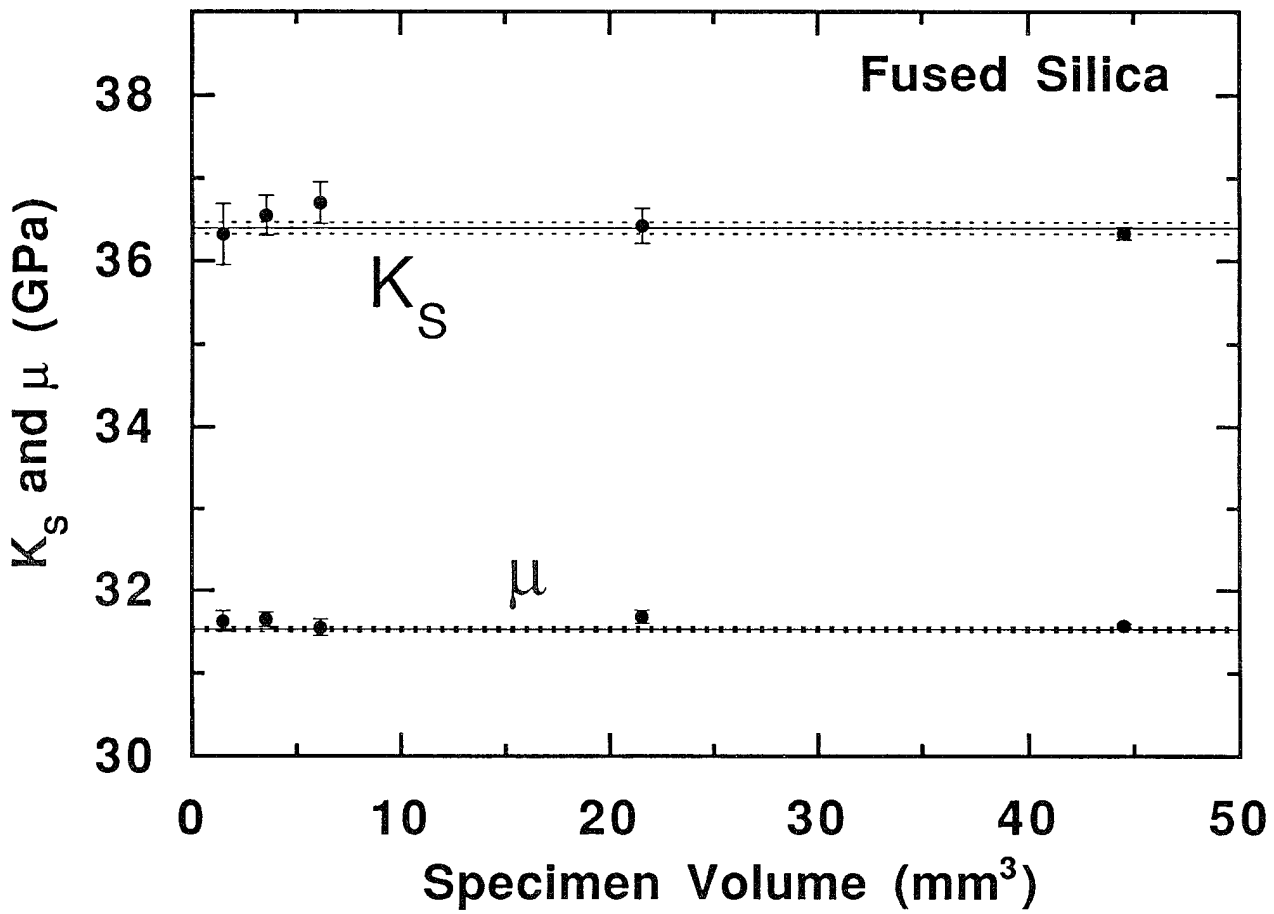


Figure 1. Specimen size effect of measured elastic moduli (RUS technique) for fused silica. Solid lines show measured values at 44.5 mm^3 for frequency extrapolated to zero gram holding mass. Dashed lines indicate errors. Data points of five specimen volumes (all at 2 grams holding force) show no systematic increase in bulk modulus, K_S, and shear modulus, μ, as volume decreases.

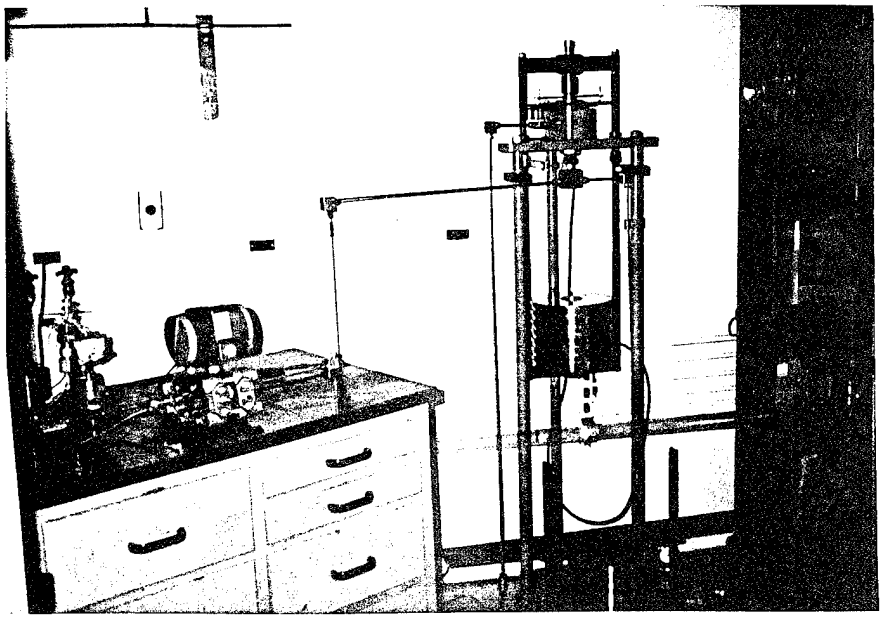


Figure 2. (a) Dead weight tester loaded to 100 bar pressure. Pressure line to left couples DWT chamber pressure to differential pressure cell (cylindrical object on table). At far left is pressure cell and specimen holding apparatus.

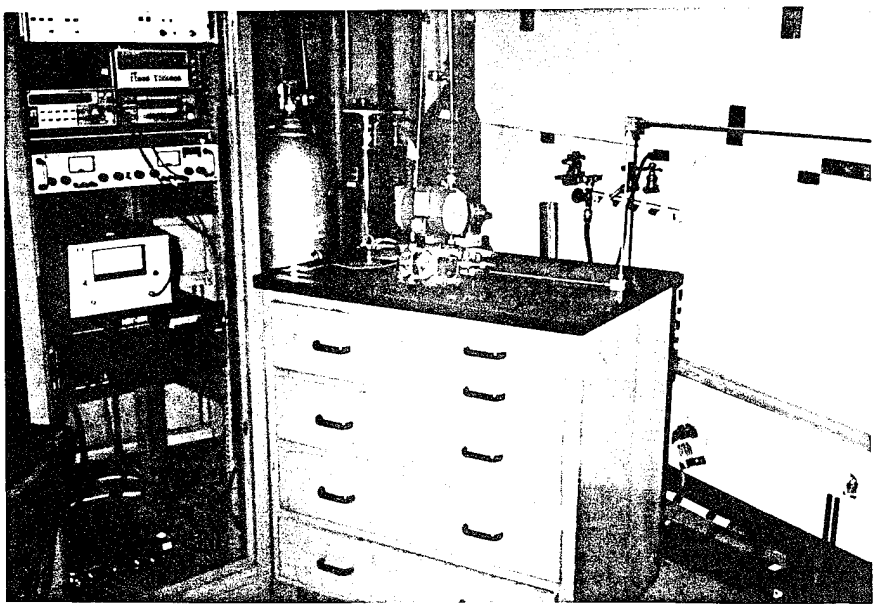


Figure 2. (b) Differential pressure cell coupled to DWT and electronics. NDI 501 and receiver box are black items in lower part of electronics rack.

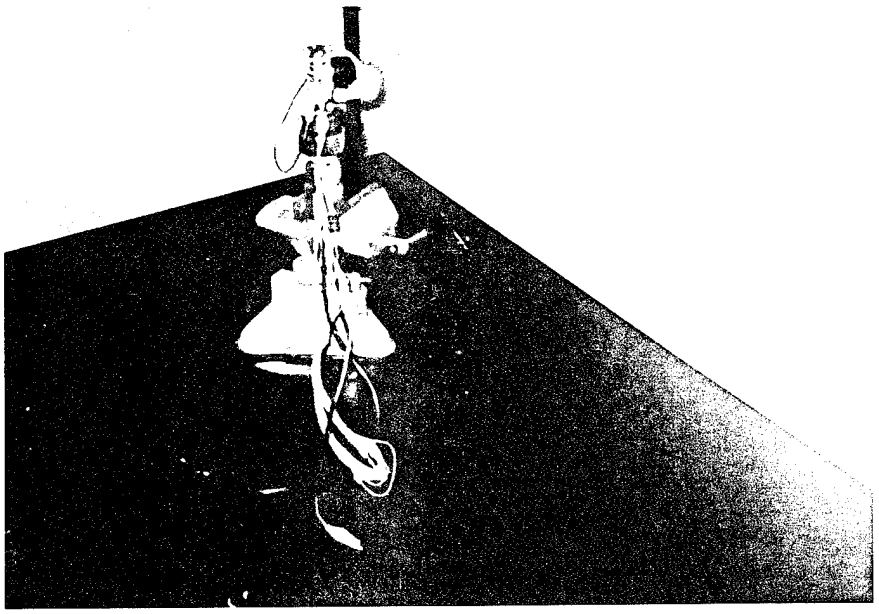


Figure 2. (c) Specimen holding apparatus. Top part inserts into pressure cell.

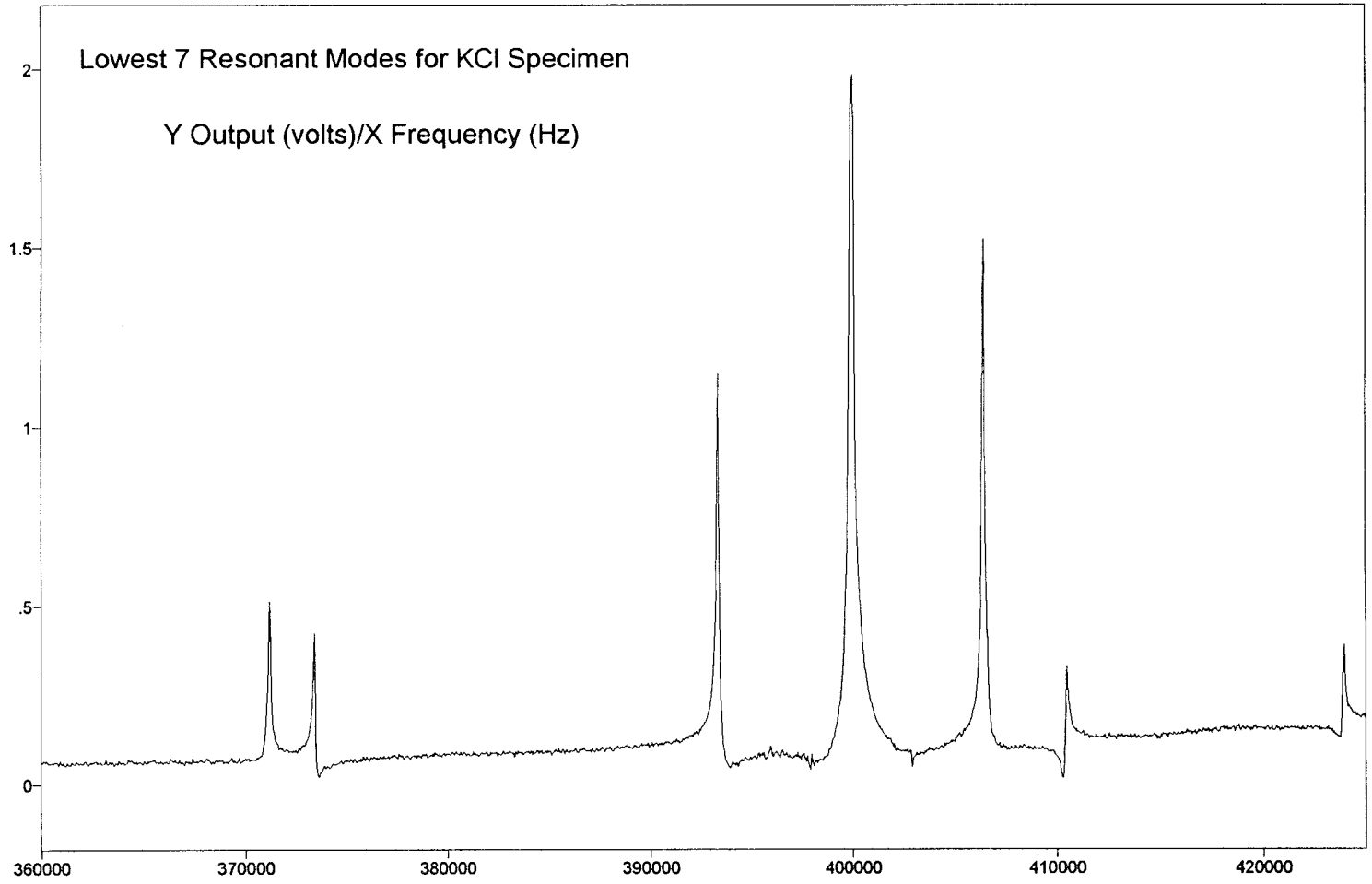


Figure 3. Sample spectrum obtained for KCl specimens using the recently constructed holding apparatus that inserts into the pressure vessel.

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